Generalized Pattern Search Methods for a Structure Determination Problem

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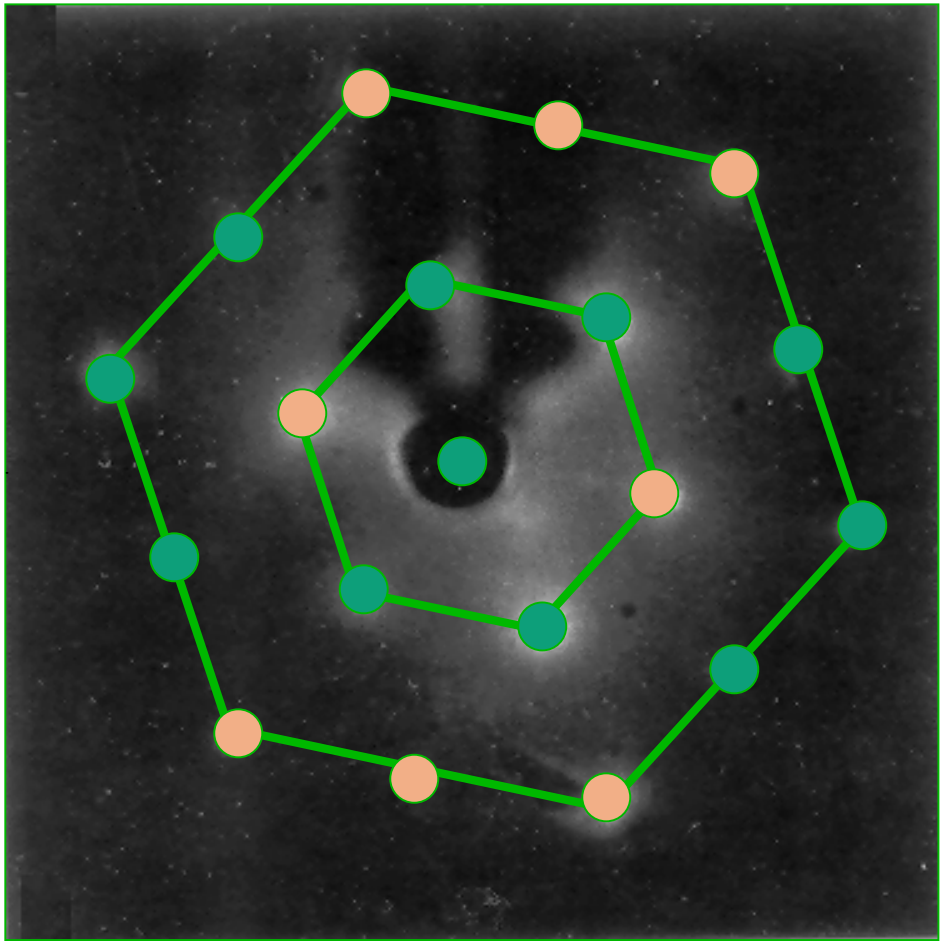
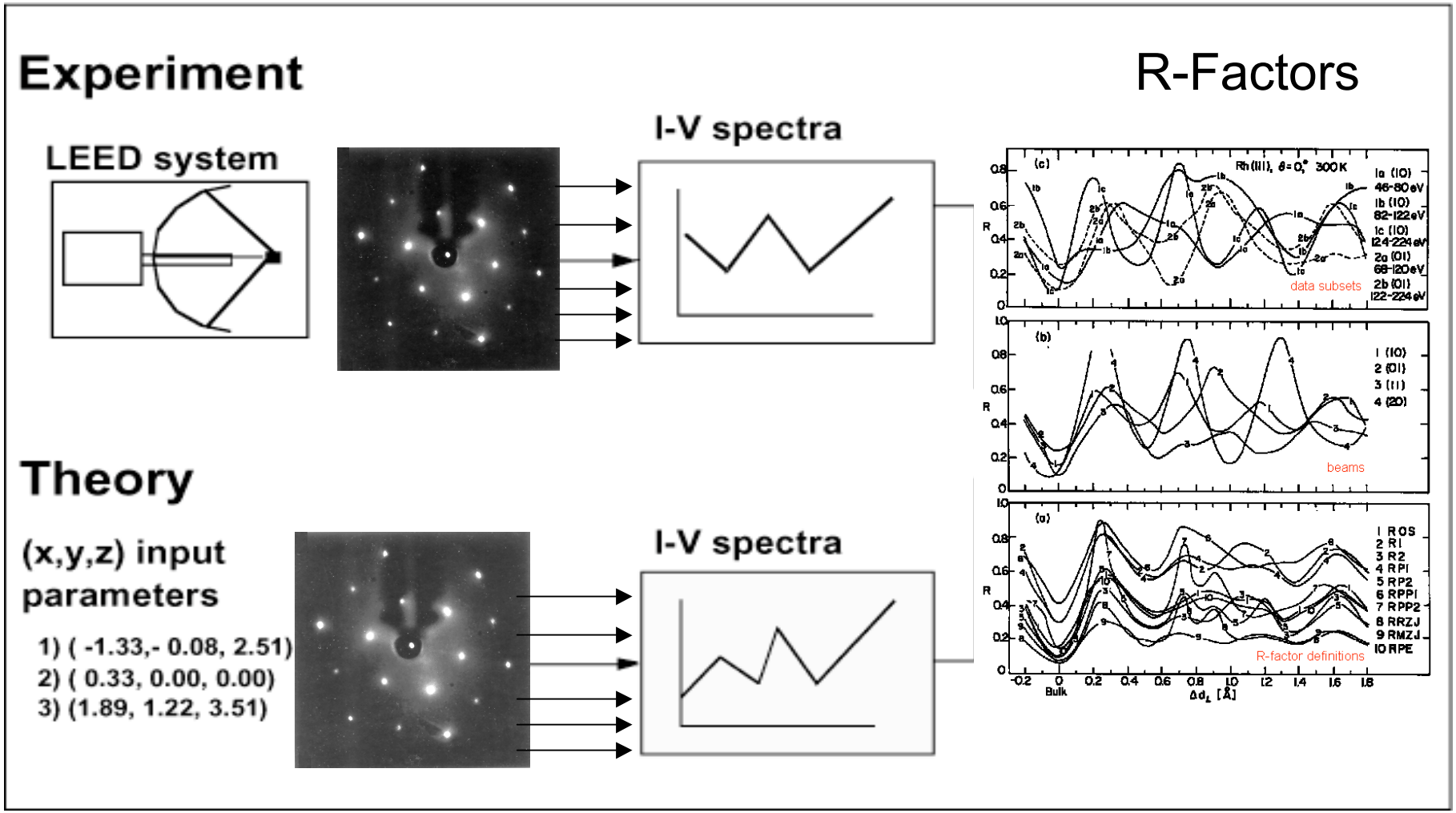
Low-energy electron diffraction (LEED)



Low-energy electron diffraction pattern due to monolayer of ethylidyne attached to a rhodium (111) surface

- Goal is to determine surface structure through low energy electron diffraction (LEED)
- Inverse problem consists of minimizing the error between experiment and theory
- Combination of local/global optimization
- Contains both continuous and categorical variables
  - Atomic coordinates
  - Ni, Li
- Function not smooth; sometimes undefined; no analytic derivatives

Low Energy Electron Diffraction

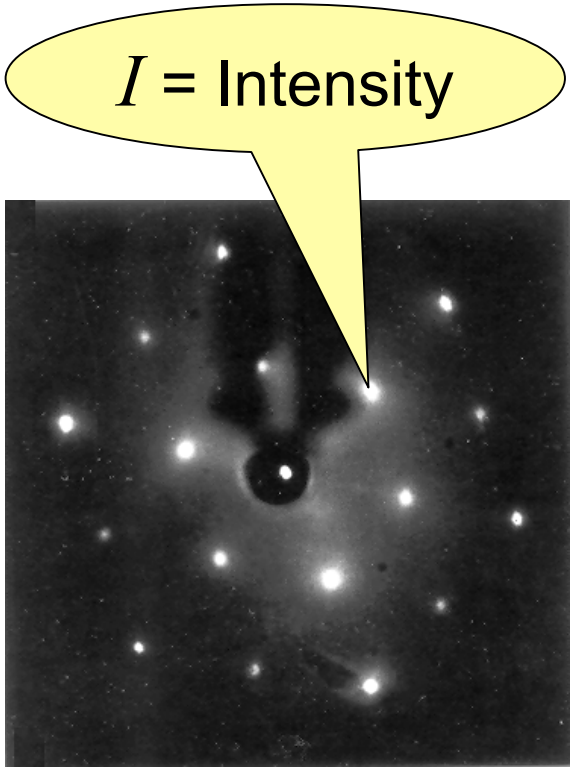


Pendry R-factor

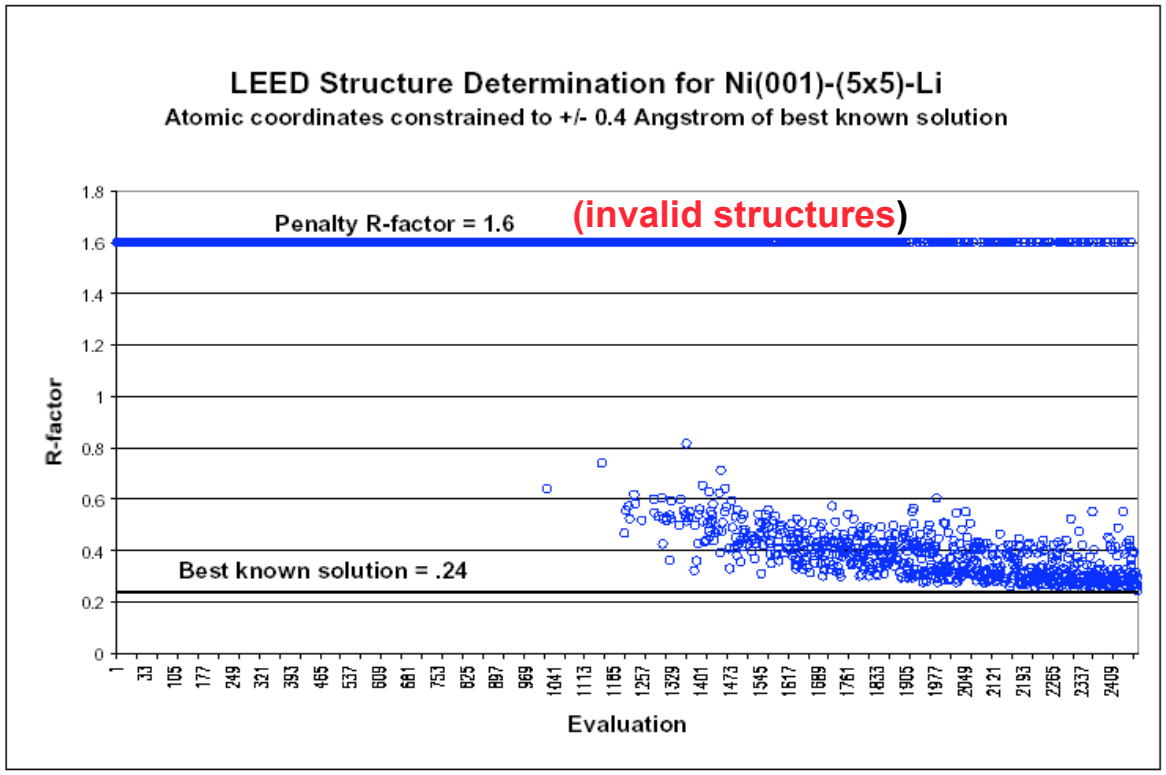
$$R = \frac{\sum_g \int (Y_{gth} - Y_{gexp})^2 dE}{\sum_g \int (Y_{gth}^2 + Y_{gexp}^2) dE},$$

$$Y(E) = L^{-1}/(L^{-2} + V_{oi}^2),$$

$$L(E) = I'(x, y, z)/I(x, y, z)$$



Previous Work

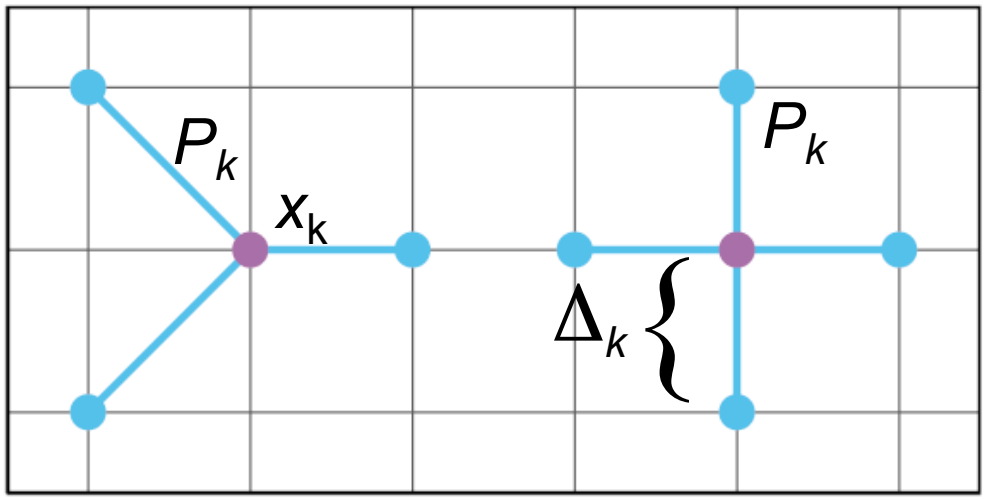


- Previous work used genetic algorithms to solve the optimization method.
- Large number of invalid structures generated.
- Overall, a solution was found - *after adding sufficient constraints*.

1. *Global Optimization in LEED Structure Determination Using Genetic Algorithms,* **R. Döll and M.A. Van Hove**, Surf. Sci. **355**, L393-8 (1996).
2. *A Scalable Genetic Algorithm Package for Global Optimization Problems with Expensive Objective Functions,* **G. S. Stone**, M.S. dissertation, Computer Science Dept., San Francisco State University, 1998.

Brief overview of pattern search methods

- Pattern search methods, Torczon, Lewis & Torczon, Lewis, Kolda, Torczon (2004), etc.
- Extension to mixed variable problems by Audet and Dennis (2000).
- Case of nonlinear constraints studied in Abramson's PhD dissertation (2002).
- Good convergence properties
- Good software available - APPSPACK (Kolda), OPT++ (Hough, Meza, Williams), NOMADm (Abramson)



Generalized Pattern Search Framework

1. Initialization: Given Δ_0 , x_0 , M_0, P_0
2. For k = 0, 1, …

   a) SEARCH: Evaluate f on a finite subset of trial points on the mesh M_k

   b) POLL: Evaluate f on the frame P_k

   Global phase can include user heuristics or surrogate functions

   Local phase more rigid, but necessary to ensure convergence

3. If successful - mesh expansion:

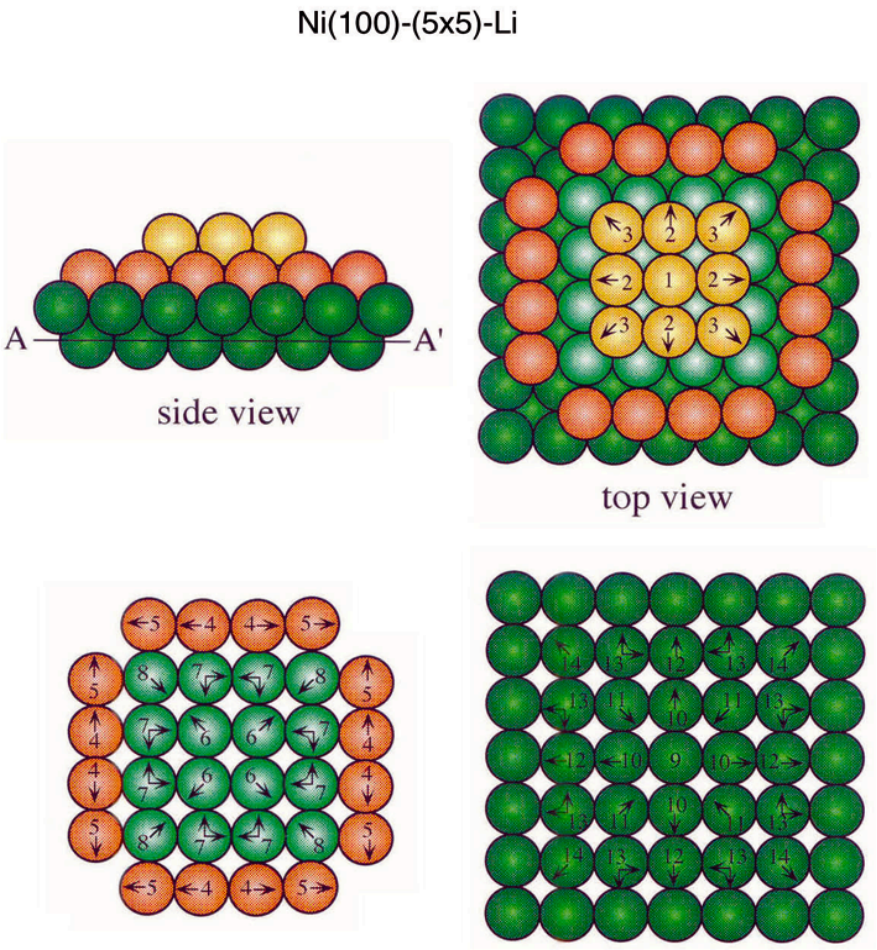
   a) $x_{k+1} = x_k + \Delta_k d_k$

4. Otherwise contract mesh

NOMADm

- Variables can be continuous, discrete, or categorical
- General constraints (bound, linear, nonlinear)
  - Nonlinear constraints can be handled by either filter method or MADS-based approach for constructing poll directions
- Objective and constraint functions can be discontinuous, extended-value, or nonsmooth.
- Available at: http://en.afit.edu/ENC/Faculty/MAbramson/NOMADm.html

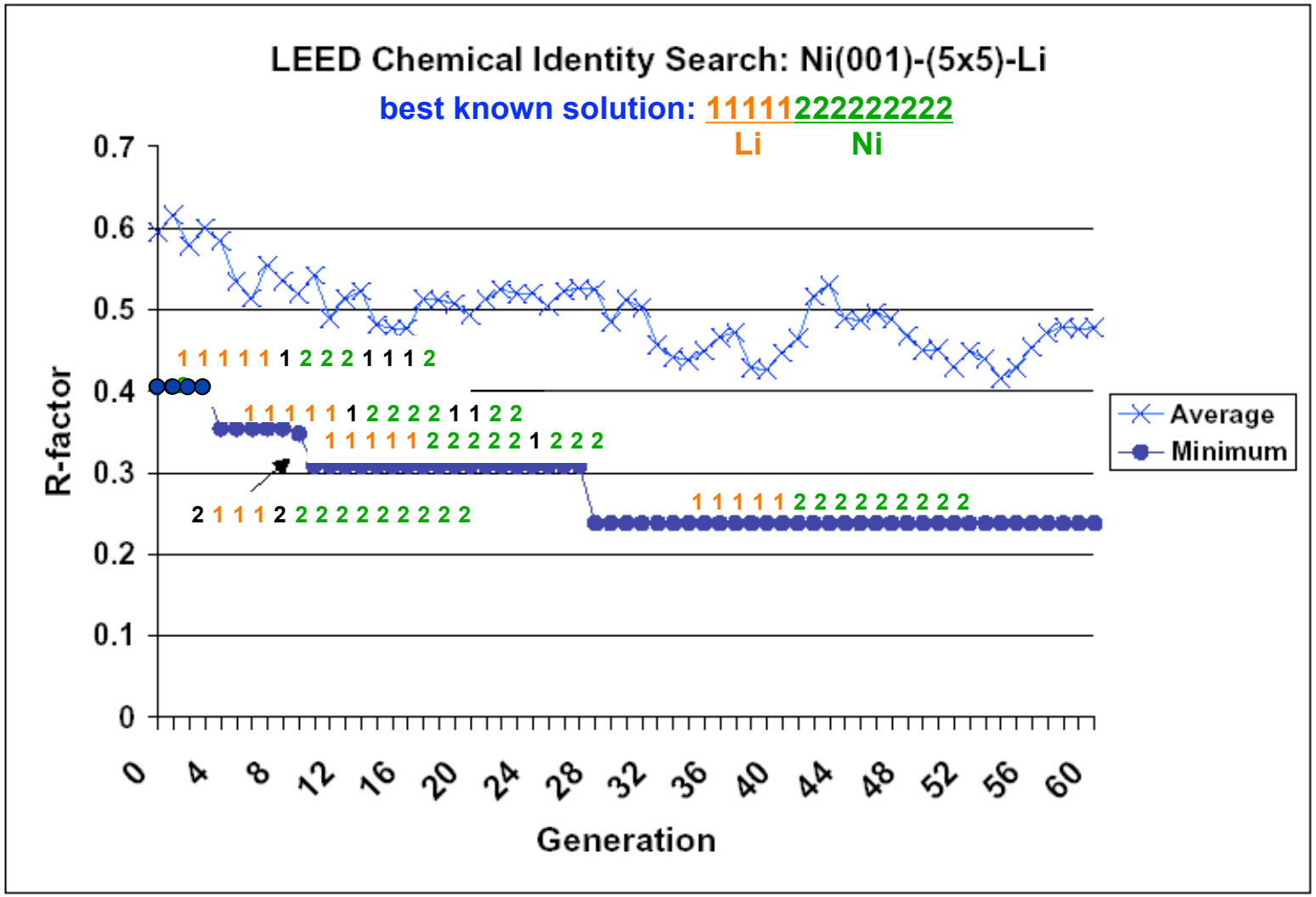
Test problem



Model 31 from set of TLEED model problems

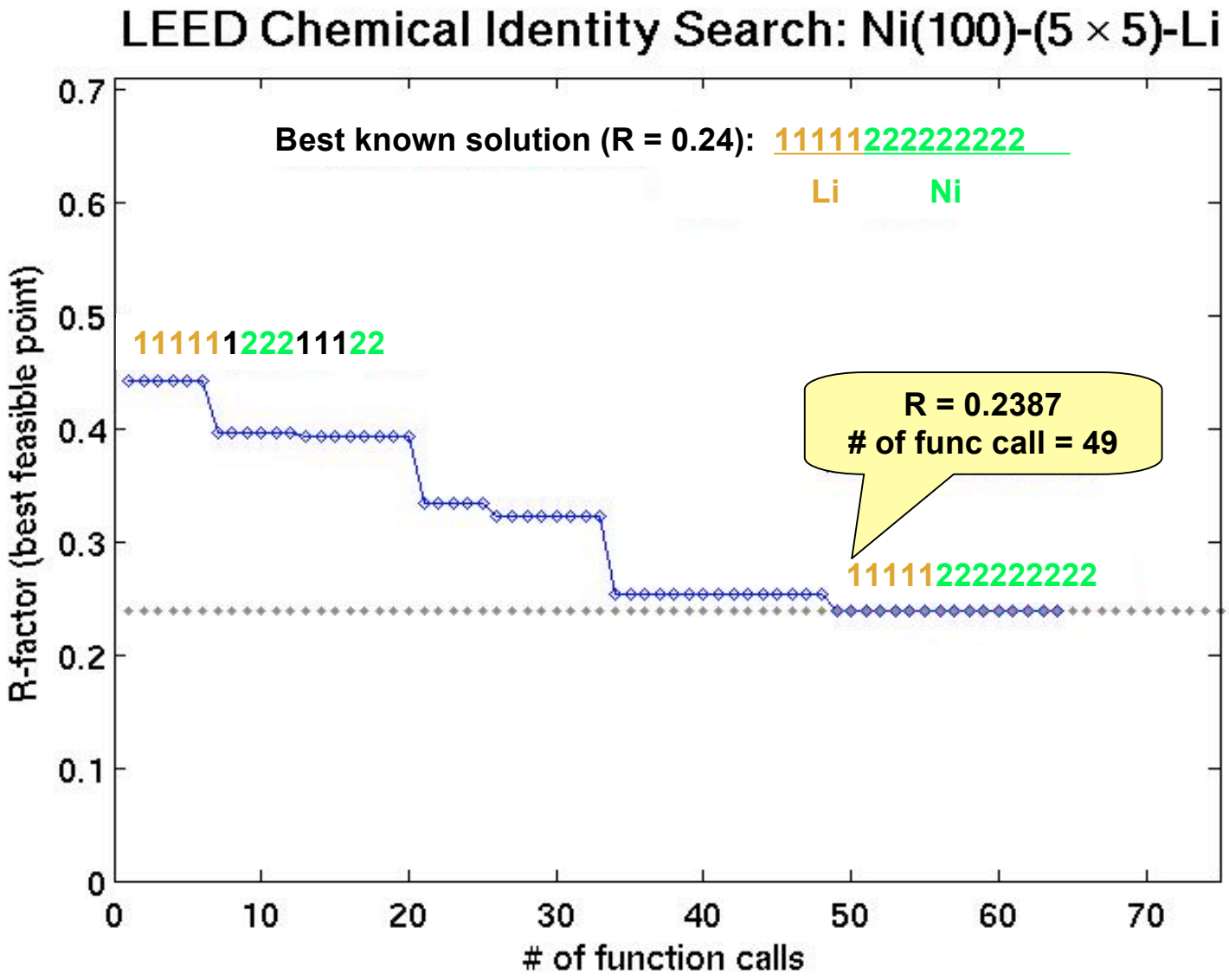
- Model contains three layers of atoms
- Using symmetry considerations we can reduce the problem to 14 atoms
  - 14 categorical variables
  - 42 continuous variables
- Positions of atoms constrained to lie within a box
- Best known previous solution had R-factor = .24

GA results - categorical variable search with fixed atomic positions

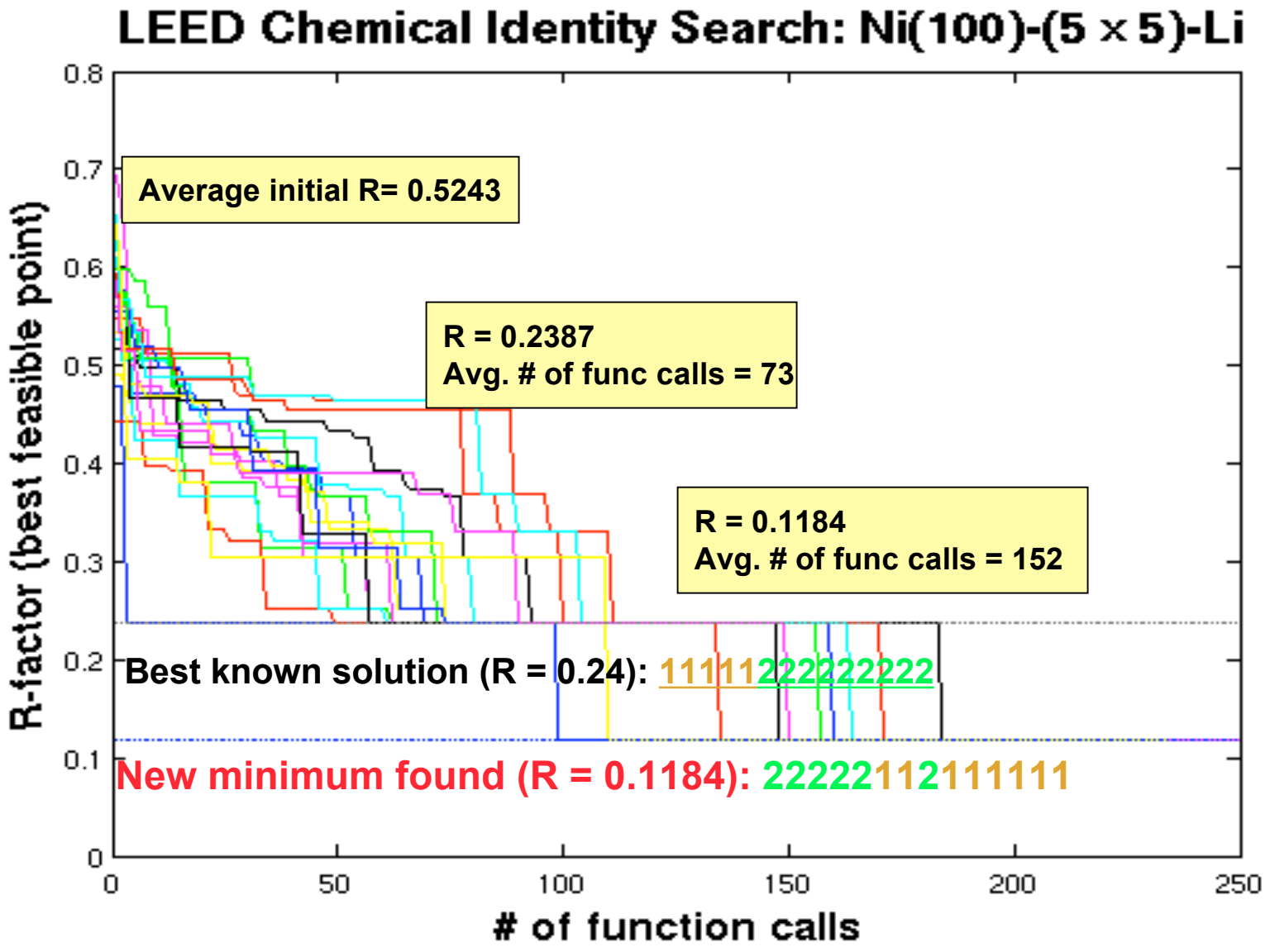


Remark: population size = 10 / Generation

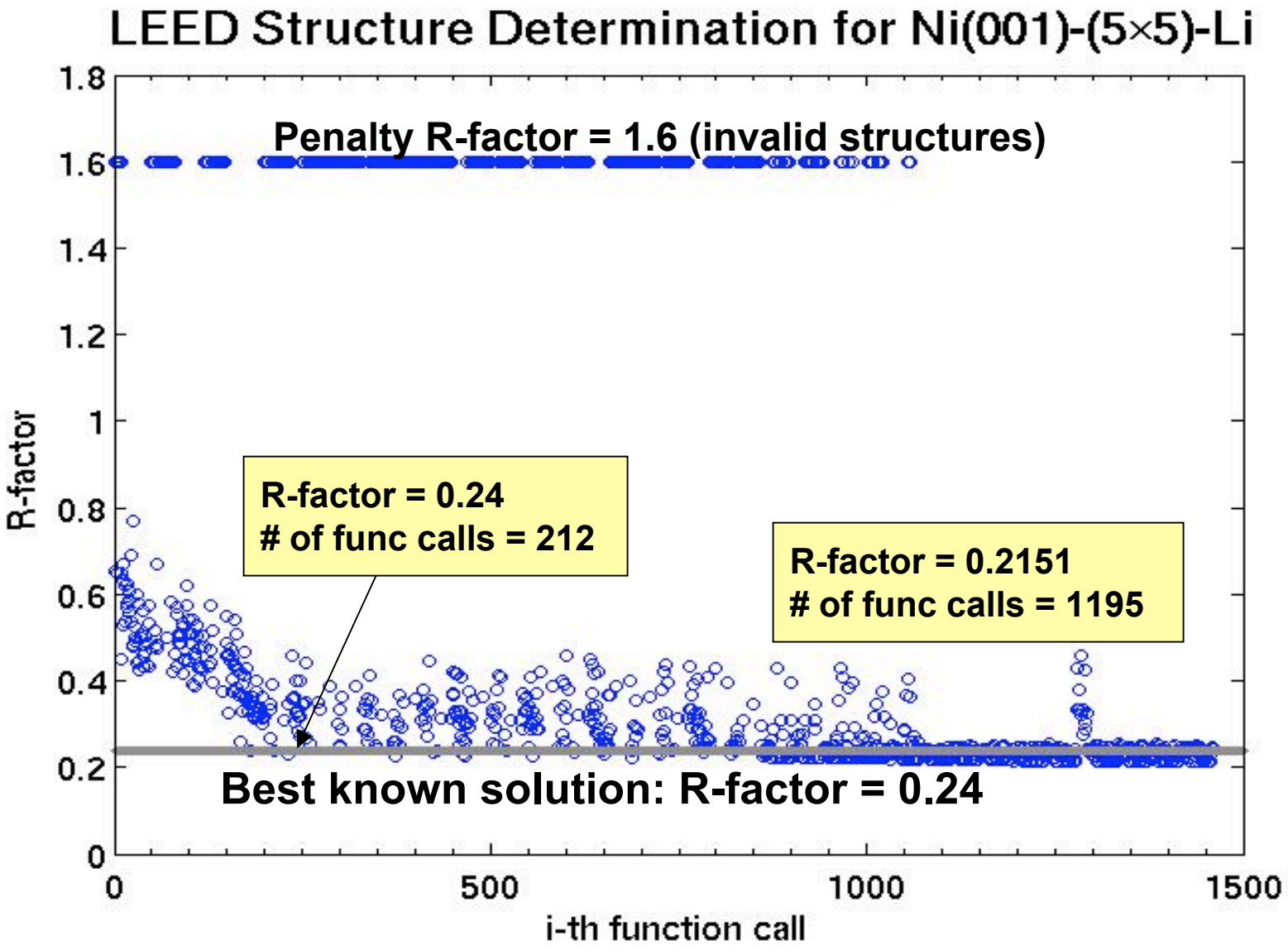
NOMAD results for categorical variables with fixed atomic positions



NOMAD results for 20 trials using LHS + GSS



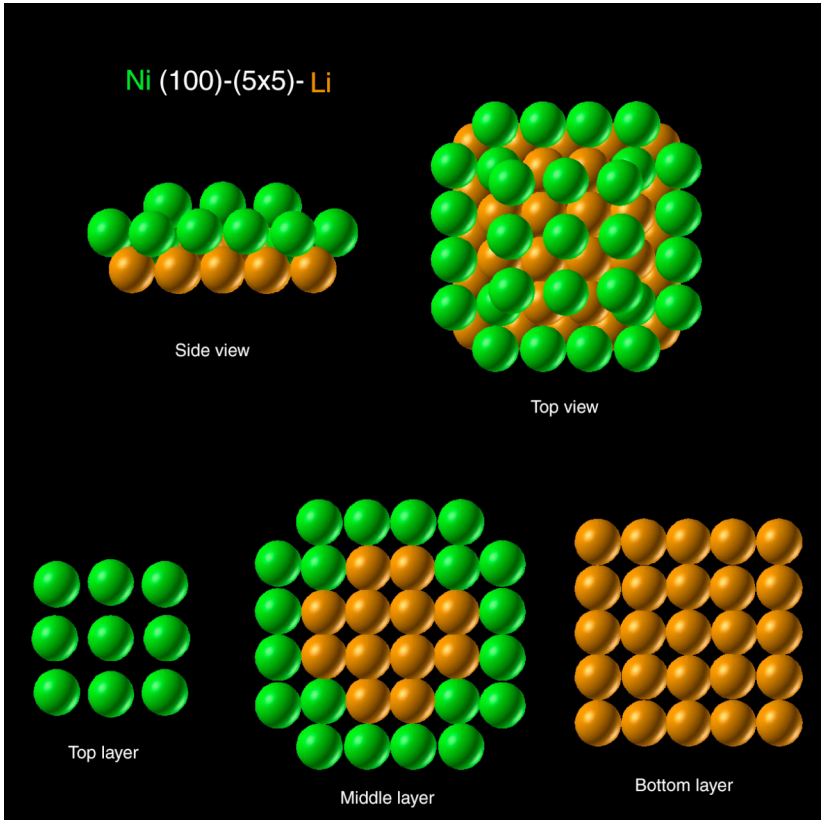
Minimization with respect to both types of variables removes coordinate constraints



LEED Chemical Identity Search: Ni (100)-(5x5)-Li

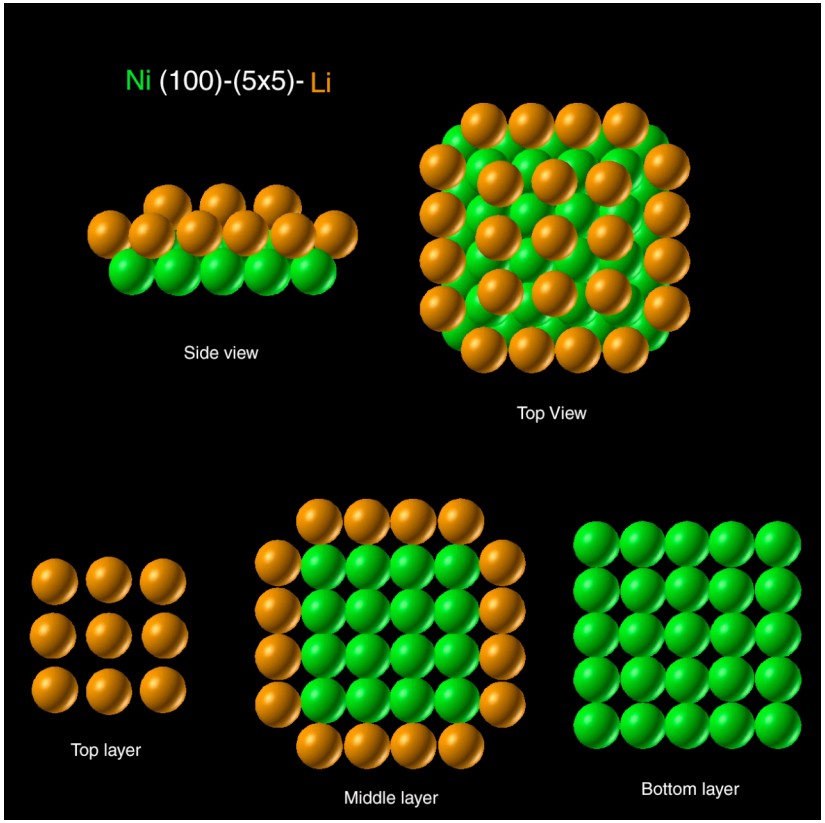
New structure found

R = 0.1184



Previous best known solution

R = 0.24



Conclusions

- Generalized pattern search methods for mixed variable problems were successful in solving the surface structure determination problem
  - On average NOMAD took 60 function evaluations versus 280 for previous solution (GA)
  - Improved solutions from previous best known solutions found in all cases
  - Generation of far fewer invalid structures
- Algorithm appears to be fairly robust, with a better structure found in all 20 trial points
- Ability to minimize with respect to both categorical and continuous variables a critical advantage for these types of problems

Acknowledgements

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Questions